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THESIS

SEARCH FOR SHORT LIVED ISOTOPES IN

VOLATILE REACTOR FUEL PRODUCTS

by

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Volatile Reactor Fuel Products

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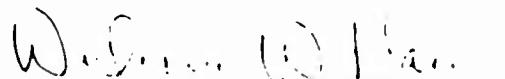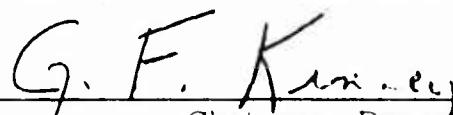
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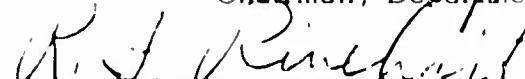
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ABSTRACT

The volatile radioactive nuclides produced by thermal neutron fissioning of uranium oxide-polyethylene fuel of the AGN-201 Reactor were collected in an evacuated aluminum cylinder. The following constituent isotopes, identified by gamma ray spectrometry, present in 20 minute old gas are; Te^{131} Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{137}, Xe^{138}, Xe^{139}, Se^{81}, Se^{83}, Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, and Kr^{89}. Radioactive daughter products identified are Cs^{137}, Cs^{138}, Cs^{139}, Rb^{88}, Rb^{89}, and I^{131}. Other isotopes of iodine, tellurium, and bromine may be present. Spectra were taken with a NaI (Th) scintillation crystal.

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LIST OF DEFINITIONS OF ABBREVIATIONS AND SYMBOLS

1. AGN-201 – The model 201 nuclear reactor produced by Aerojet General Nucleonics Corporation, San Ramon, California

2. MEV – million electron volts

3. NPS – The Naval Postgraduate School, Monterey, California

4. Elemental symbols:

    Al – aluminum
    Au – gold
    Br – bromine
    Co – cobolt
    Cs – cesium
    I   – iodine
    Kr – krypton
    Mn – manganese
    Rb – rubidium
    Se – selenium
    Te – tellurium
    U   – uranium
    Xe – xenon

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I. INTRODUCTION

In the past many attempts have been made to analyze both
qualitatively and quantitatively the gaseous effluents given off from
the core of the AGN-201 Nuclear Reactor at the Naval Postgraduate
School. Bredderman [1] conducted a detailed analysis of the gamma
ray spectrum of the gas given off by the core of the reactor and sampled
two hours after peak of power operation. The principle radioactive
isotopes present based on gamma ray photopeak energies and half
lives were found to be Kr^{85m} 10.2%, Kr^{87} 9.0%, Kr^{88} 47.4%, Xe^{133}
18.6%, Xe^{135} 4.4%, and an unidentified activity 10.4%. He observed
the total activity to be about 363 microcuries per milliliter. Laskaris [2]
analyzed the reactor core gas by means of a gas chromatograph and
found the gas to be comprised almost 50% by volume of hydrogen, with
the remaining constituents being methane, ethane, ethylene, and other
light saturated, unsaturated, and cyclic hydrocarbons. He stated the
oxygen, carbon dioxide, and carbon monoxide detected in these gases
was directly attributed to the impurities of the nitrogen gas used to
pressurize the reactor. At the same time he made an activity analysis
that demonstrated the existence of xenon and krypton radio-nuclides
in the effluent gases. He further determined the percentage by volume
of krypton and of xenon to be less than 0.5% each. In a later work by
Cole [3] the gases produced by radiation of the polyethylene moderated
uranium oxide fuel were analyzed for both radioactive and nonradioactive

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species. He showed the hydrogen content to be about 60% by volume, the ethane-ethylene to be 17%, methane 19% with the remaining composition being that of the higher alkanes and cyclic hydrocarbons. His gas chromatograms showed xenon to be faintly detectable. Trace quantities of krypton and xenon were successfully separated without isotopic dilution using gas-solid chromatography. The radioactive species identified by Cole were Kr^{85m}, Kr^{87}, Kr^{88}, Xe^{133m}, Xe^{133}, Xe^{135}, Xe^{138}, I^{131}, Cs^{138}. Cole believed that the iodine present would be in the form of hydrogen iodide or compounds such as methyl iodide and ethyl iodide. However, it should be pointed out that no hydrogen iodide or methyl iodide was observable in his chromatograms. Cole further indicated the possible existence of Rb^{88}, Rb^{89}, Xe^{131}, Xe^{135m}, I^{133} and I^{135} based solely on photopeak identification with no supporting half life data. At the same time Lindsay [4] conducted a very thorough half life study of various photopeaks found in the gamma ray spectrum of the volatile reactor fuel products. He observed some 34 to 40 photopeaks of which several were not strong enough to make any detailed half life study. He did identify 11 constituent nuclides including two radioactive daughters by this method. They were I^{131}, Xe^{131}, Xe^{133m}, Xe^{133}, Xe^{135}, Xe^{138}, Kr^{85m}, Kr^{87}, Kr^{88}, Cs^{138}, and Rb^{88}. He further reported the possible presence of I^{133}, I^{134}, I^{135}, I^{136}, Xe^{137}, Kr^{89}, and Rb^{89} from a photopeak energy evaluation. Lindsay proposed the presence of Br^{84m}, and Br^{84} isotopes. These

bromine isotopes have half lives of 6 and 32 minutes respectively.
This suggestion plus that of the possible presence of I^{136}, which has
a half life of 86 seconds, required an extremely rapid method of ex-
traction of the gas be developed before a positive identification could
be made.

The thermal neutron fissioning of uranium 235, has been
thoroughly investigated by numerous people and a fine compilation of
fission product yields has been prepared by Katcoff [5]. The most
probable masses of the fission fragments from a thermal neutron fission
reaction are 93 and 134 atomic mass units with 7 and 8% fission yields
respectively. Masses which have fission yields greater than 3% are 88
through 103 and 131 through 146. Most of the fission fragments are
metallic atoms although nuclides of xenon, iodine, krypton, and bromine
are chemically stable in gaseous form. Therefore, it is safe to assume
that any gaseous products from a fission reaction should likely contain
isotopes of these elements. Furthermore, the identification of one
isotope of an element as a constituent in the reactor gas implies the
other fission product isotopes of that element are also present. This
leads to a mixture of isotopes of one or more elements whose spectrum
may be difficult to resolve in areas where different gamma rays have
similar energies.

Photopeaks are assumed to have from one to six decays of
different half lives. This assumption is based on the number of nuclides,
with prominent gamma ray energies equal to energies included in the

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photopeak, believed to be present in the reactor gas. A positive identi-
fication of an isotope is made when the half life of that isotope is
observed for one of the strong photopeaks of that isotope. Daughter
products can be identified by observing the growth of a strong photopeak
and then following the decay of that photopeak. A computerized program
for the processing of radioactive decay data and analyzing the decay
curve for up to 10 components in the curve has been prepared by
Rogers [6].

II. EXPERIMENTAL

A.   PRODUCTION OF GASES

The gaseous products from the fissioning of the uranium oxide-
polyethylene mixture were generated and trapped in an aluminum
cylinder and quartz glass vial arrangement similar to that shown in
Figure 1. Two fuel discs, about one centimeter thick and 4.5 centi-
meters in diameter, were placed inside an aluminum cylinder. Each
fuel disc is a mixture of 4 grams of UO_2, with 20% enrichment in U^{235},
and 100 grams of polyethylene. The cylinder is sealed at one end by a
neoprine O-ring seal and a bellows valve is attached to the other end.
The cylinder is then evacuated to a pressure of less than 1 micron.
The glass vials were then attached by means of a standard taper joint
to the valve and the valve then opened. This introduced a small
quantity of air into the cylinder and was useful in diluting the sample
for counting purposes. The pressure inside the cylinder and glass vials
was found to be approximately 200 microns. The cylinder and the glass
vials were then encased in carbon blocks and placed in the access port
of the AGN-201 Reactor near but outside of the core. The reactor was
operated at a power of 50 watts for 15 minutes subjecting the fuel
elements to a thermal neutron flux of approximately 10^9 neutrons per
square centimeter per second. Zero time for decay counting purposes
was defined as the time at which the reactor was shut down, however,
it must be pointed out that the composite age of the gas formed in the

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cylinder was from five to twenty minutes old at the start of counting. Even so this method produces a more definite sample of gas than was obtained by previous workers, Bredderman and Laskaris. Their methods of extracting gas from the core tank of the reactor through a copper tubing and manifold system resulted in a sample which contained gas evolved from different operations of the reactor. This gas could not be said to have a specific age as the gas composition would have varied from sample to sample.

B.   FRACTIONATION AND EXTRACTION

Upon shut down of the reactor the cylinder was removed and the bottom vial was placed in a cooling bath composed of a slurry of dry ice and trichorolethylene. After one minute of cooling the vial was sealed and removed to the scintillation counter. The upper vial was then placed in a liquid nitrogen cooling bath and cooled for several minutes before being sealed and counted. Several runs were made later in which the gas sample vial received no cooling but was sealed as soon as possible, permitting counting within 2.5 minutes of shut down.

C.   MEASUREMENT OF RADIOACTIVE SPECIES

Gas samples were placed in the well of a 3" by 3" sodium iodide scintillation crystal which is coupled to a photomultiplier tube. The resulting impulses of electrical energy are fed into a 512 channel analyzer. The multichannel analyzer was calibrated by utilizing the

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prominant photopeaks of Cesium 137 (0.032 MEV and 0.662 MEV),

Gold 198 (0.069 MEV and 0.411 MEV), Manganese 54 (0.840 MEV),

Cobalt 60 (1.17 MEV and 1.32 MEV), and Sodium 24 (1.370 MEV and

2.753 MEV). The electrical pulses of varying heights which correspond

to various energies, are then counted and stored in the memory unit of

the multichannel analyzer. The associated equipment with this analyzer

includes an oscilloscope for the visual display of stored data, a tele-

typewriter for printout of total number of counts in each channel, a

mechanical plotter for plotting the gamma ray spectrum and a tape

punch mechanism for permanent recording of the data. This tape can

be fed back into the multichannel analyzer at will for spectrum com-

parison studies.

Two single channel analyzers were employed to follow the decay

of prominent gamma photopeaks. The signal from the photomultiplier

tube is passed through a preamplifier and attenuator circuit before

reaching the discriminator circuit of the scaler. The discriminator can

be adjusted so that an energy window is formed to allow specific pulse

height signals to pass to the counter. The single channel analyzers

were calibrated in the following manner. First, appropriate calibration

of the 512 channel analyzer was made utilizing the standard test samples.

Then a spectrum from the previous experiment was fed into the memory

core of the 512 channel analyzer. An appropriate energy window was

selected from the display on the oscilloscope. A signal from a variable

height pulse generator was fed into the multichannel analyzer at the

same point that the signal from the photomultiplier tube enters. The pulse generator is then adjusted so that its signal is counted on the channel corresponding to one edge of the photopeak window. This signal, equaling a specific gamma ray energy, is fed into the pre-amplifier and attenuated to allow counting in the best operating region of the single channel scalers. The appropriate discriminator is adjusted to count this pulse. As a result pulses whose heights fall within the photopeak energy window are counted. Counting intervals with the single channel analyzer varied from 30 seconds to 10 minutes as appropriate to the half lives of the isotopes estimated in the photo-peak. The length of the counting period or experiment usually ran for 300 minutes after shut down of the reactor, although counts were taken as late as 1000 to 1500 minutes after shut down, in order to establish a long term decay component. The photopeaks that were followed by this method were the 0.157, 0.201, 0.224, 0.267, 0.410, 0.455, 0.585, and 1.745 MEV as these were the photopeaks that were strong and showed some indication of short half life components of less than 60 minutes.

D. CHEMICAL ABSORPTION TECHNIQUES

A chemical separation was attempted by bubbling the reactor gas through a solution of sodium sulfite to absorb and insure reduction of the iodine and bromine into their lowest oxidation states. The gas was pumped from the cylinder, located inside the reactor, through three

traps connected in series. The entire system was evacuated initially. A solution containing 140 milliliters of 1$\underline{M}$ sodium sulfite and 10 milliliters of 1$\underline{M}$ hydrochloric acid was placed in the first trap. The second and third traps were surrounded by dewar flasks containing liquid nitrogen. The reactor was run at 50 watts for 15 minutes. After shut down the gas was allowed to bubble through the sodium sulfite solution by action of the vacuum pump for 10 to 15 minutes. A quick check with a geiger counter indicated most of the activity had been trapped in the middle trap containing liquid nitrogen. A very small amount of radiation was detectable in the last trap. The sulfite solution was placed in a beaker and counted. The high activity species in the second trap was transferred into an evacuated glass container and also counted.

III. RESULTS

In general the results of this experiment are best given in
tabular form or in reproductions of the gamma ray spectra. Reactor gas
was produced on twelve separate runs. Three of these runs were for the
purpose of obtaining gamma ray spectra only and no significant half
life data were measured. Eight successful runs were made for half
life determination of photopeaks. One attempt was made to follow the
decay of the 0.032 MEV photopeak but results were inconclusive since
numerous isotopes have minor gamma and x-ray energies in this region
and their resolution is poor. Readings were erratic because of the
highly complex decay, growth of daughter products, and Compton
scattering from higher energy photons. Gamma ray spectra were also
taken during these runs and in all some 174 spectra were obtained.
Listed in Table I are the photopeaks and their respective energies.
In general the spectra obtained were very similar to those reported by
Lindsay with some variations of the energy values. Energies reported
by Bredderman are from gas of an indeterminate age which includes
daughter products. Energies reported by Lindsay are from gas produced
in a similar method described in this report and also includes daughter
products. It was found that the multichannel analyzer did not maintain
a linear ratio of energy per channel throughout all the channels. In-
stability in the calibration by as much as 2 channels per 24 hour period
was seen. It was observed that the calibration of the multichannel

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analyzer holds quite well for a 3 or 4 hour period during the day, however at night drifting occurred. The single channel analyzers did not maintain their calibration as well as hoped. At the completion of runs the same calibrated signals from the pulse generator were fed back into the scalers and it appeared that the discriminators had shifted. The pulse signal generator held its calibration quite well. When a signal was fed from it into the multichannel analyzer no detectable deviation of that signal into adjacent channels could be observed. This was true at the beginning and at the end of a run.

A.   EARLY GAMMA RAY SPECTRUM ANALYSIS

Shown at Figure 2 are comparison spectra, 0 to 1 MEV range, of the dry ice cooled vial and liquid nitrogen cooled vial. It was hoped that cooling with dry ice would give a partial fractionation or enrichment of the iodine and bromine isotopes with a resulting smaller amount of krypton and xenon isotopes. Cooling at liquid nitrogen temperature freezes out all of the gaseous isotopes. The significant difference between the dry ice cooled and the liquid nitrogen cooled samples is evident in the 0.455 MEV photopeak which is slightly higher in the liquid nitrogen cooled spectrum. Figure 3 are comparison spectra, 0 to 2 MEV range, between a vial that was cooled with dry ice and a vial that received no cooling at all. No significant differences were observed in comparable spectra between dry ice cooled gas samples and the gas samples which received no cooling. Comparison

spectra between a dry ice cooled gas sample and the chemical absorbed sample are shown in Figure 4. Of significance here is the absence of the prominent photopeaks at 0.155, 0.201, 0.263, 0.432 MEV. A significant change had occurred about an hour later in the doublet photopeak 0.416 and 0.470 MEV. The spectrum taken of the activity trapped in the liquid nitrogen bath was essentially the same as corresponding spectra from a dry ice cooled vial. Figures 5 and 6 are 0 to 2 and 0 to 4 MEV range spectra taken during the 9th run. The decay of the prominent photopeaks is readily apparent and will be discussed in the next paragraph. The growth of certain photopeaks is also noticeable and will be commented upon later.

B.   HALF LIFE ANALYSIS

The decays of nine separate photopeaks were followed by the single channel analyzers and used as input data for the "FRANTIC" program. The 0.032 MEV photopeak showed so much change, both increasing and decreasing, that no useful half life measurement could be made. Other decay measurements are summarized in Table II for each photopeak. The "FRANTIC" program corrects the observed data for background, dead time of the counting system, and the decay from zero time. The program then goes through a least squares fit of the data to obtain the decay curve. Included as input parameters to the computer program are the decay constants for the suspected isotopes in the photopeak. These decay constants are not held fixed by the computer

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program but are allowed to vary by a factor of three per iteration to obtain the best fit of the curve. Up to fifty iterations are allowed to find the best fit. Calculations were made to fit each curve with a minimum of two components and as many as six components. Various combinations of input decay constants were tried to determine if certain combinations fit better than others. The "FRANTIC" program is designed to accommodate up to 10 components in a decay curve, however, it seems to have difficulty separating components whose half lives differ by less than a factor of three. Several attempts were made to compute decay constants graphically from the corrected data. These decay constants were then used as input into "FRANTIC" to determine a more accurately fitted curve to the set of data points. In some cases the same fit was obtained but with fewer iterations. In other cases a poorer fit was obtained and in some cases a better fit to the curve was obtained. The "FRANTIC" program computes a standard deviation for each data point from the calculated decay curve. The total number of data points used for the calculation and the number of data points deviating more than two standard deviations are indicated in Table II. The weighted mean variance for each curve is also given. The smaller the weighted mean variance the better is the fit of the curve. A weighted mean variance of one is considered a very good fit to a set of experimental data.

GAMMA PHOTOPEAKS IN EARLY SPECTRA OF REACTOR FUEL GAS

| Photopeak Number	Energy (MEV)	Previously Reported
1	0.032 ± 0.008	[1] [4]
2	0.155 ± 0.008	[1] [4]
3	0.201 ± 0.008	[4]
4	0.224 ± 0.008	
5	0.263 ± 0.008	[4]
6	0.316 ± 0.016	[1] [4]
7	0.408 ± 0.008	[1] [1]
8	0.432 ± 0.016	[4]
9	0.455 ± 0.008	[4]
10	0.524 ± 0.008	[4]
11	0.539 ± 0.008	[4]
12	0.585 ± 0.008	
13	0.613 ± 0.016	[4]
14	0.654 ± 0.016	[4]
15	0.854 ± 0.008	[1] [4]
16	1.023 ± 0.008	[4]
17	1.231 ± 0.008	[1] [4]
18	1.392 ± 0.016	
19	1.423 ± 0.016	[1] [4]
20	1.496 ± 0.032	
21	1.745 ± 0.008	[4]
22	1.841 ± 0.016	[1] [4]
23	1.999 ± 0.016	[4]
24	2.203 ± 0.016	[4]
25	2.344 ± 0.016	[4]
26	2.392 ± 0.016	[1] [4]

TABLE I

HALF LIVES OBSERVED

Observed Half Life in Minutes

Observed Photopeak	Run	1st Component	2nd Component	3rd Component	Weighted Variance of Fit	Number of Points	Points Deviating 2	Number of Iterations
0.155 MEV	D	2.83	18.2	227.4	0.811	105	3	9
0.201 MEV	E	0.56	5.13	127.0	1.154	100	6	26
0.224 MEV	J	3.39	22.1	131.5	2.799	160	32	12
0.263 MEV	C	2.38	17.6	----	1.098	34	2	10
	D	3.49	15.2	140.2	1.065	125	3	15
	G-1	4.6	15.0	160.0	0.878	110	2	31
	G-2	3.29	15.0	215.0	0.670	110	3	22
0.408 MEV	E	2.70	14.3	75.1	1.03	105	5	13
0.455 MEV	H-1	2.49	25.6	123.6	2.147	110	5	10
	H-2	2.63	29.9	129.4	1.549	110	5	15
0.585 MEV	I	2.56	13.1	62.3	1.104	90	5	16
1.745 MEV	K-1	2.56	81.5	-----	1.56	130	15	10
	K-2	2.74	109.0	-----	2.167	140	11	26

TABLE II
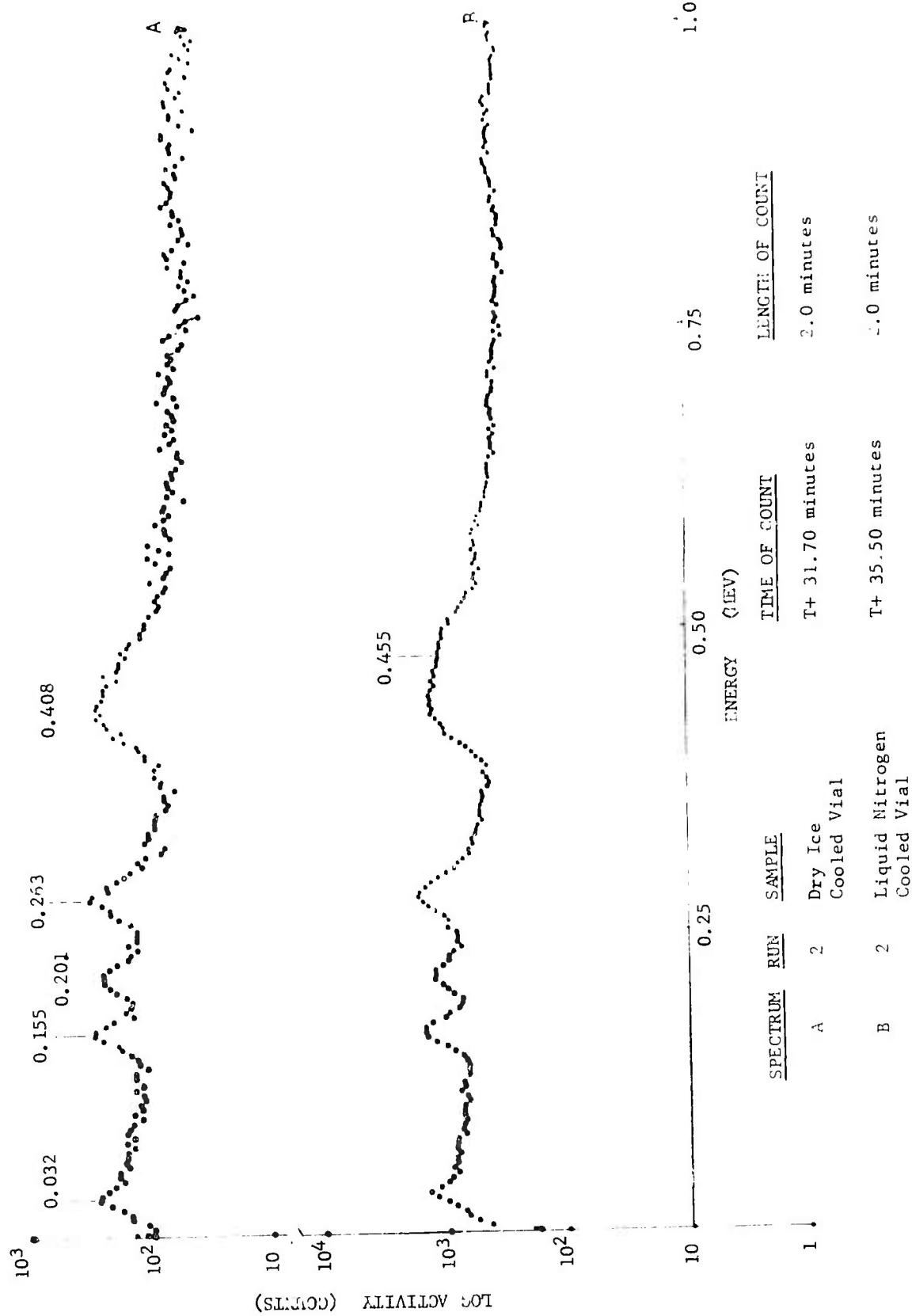
Comparison Spectra of Dry Ice and Liquid Nitrogen Cooled Gas Samples.

Figure 2

SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
A	2	Dry Ice Cooled Vial	T+ 31.70 minutes	2.0 minutes
B	2	Liquid Nitrogen Cooled Vial	T+ 35.50 minutes	2.0 minutes
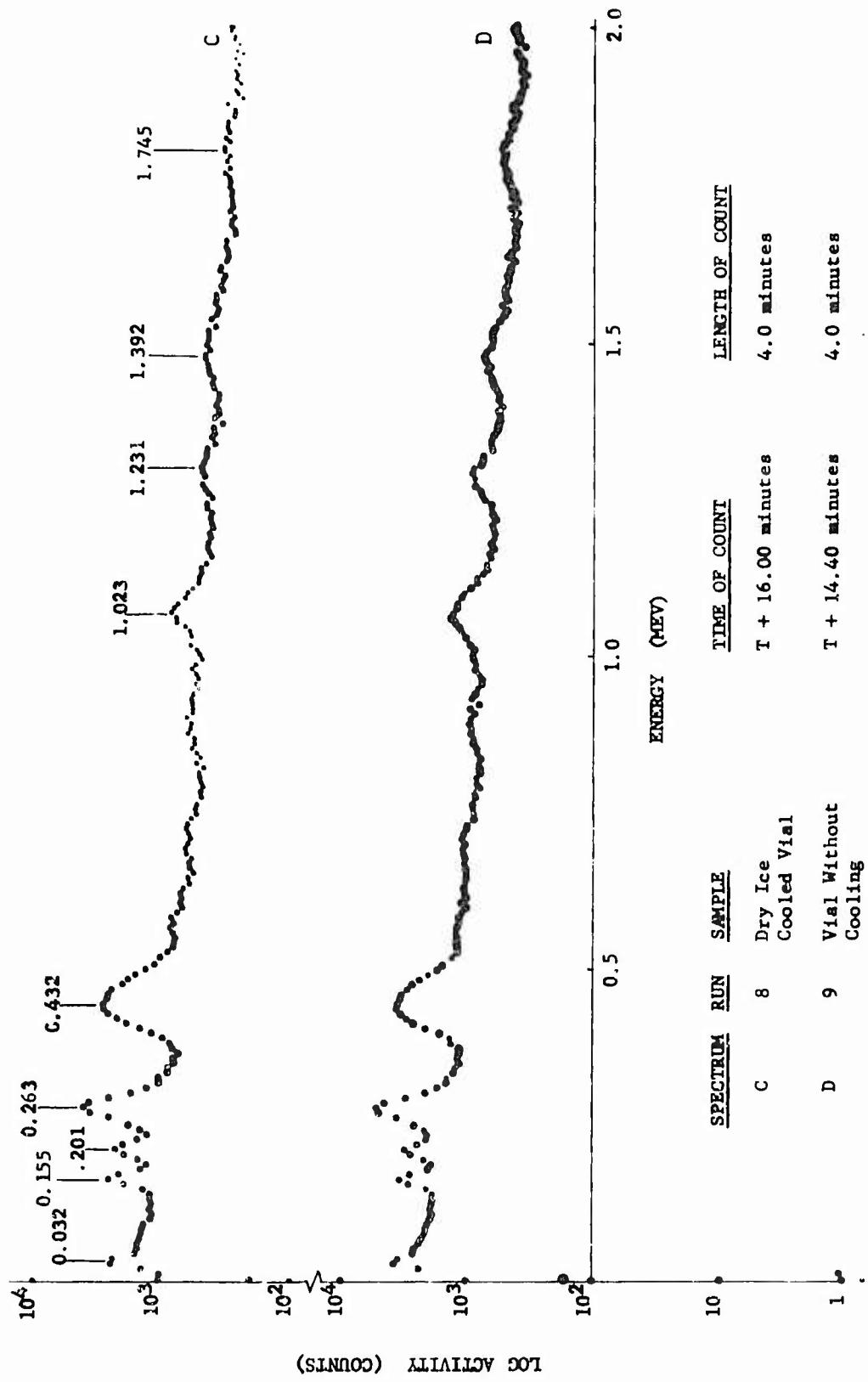
Comparison Spectra of Dry Ice Cooled Gas Sample and Sample Without Cooling.

Figure 3

SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
C	8	Dry Ice Cooled Vial	T + 16.00 minutes	4.0 minutes
D	9	Vial Without Cooling	T + 14.40 minutes	4.0 minutes

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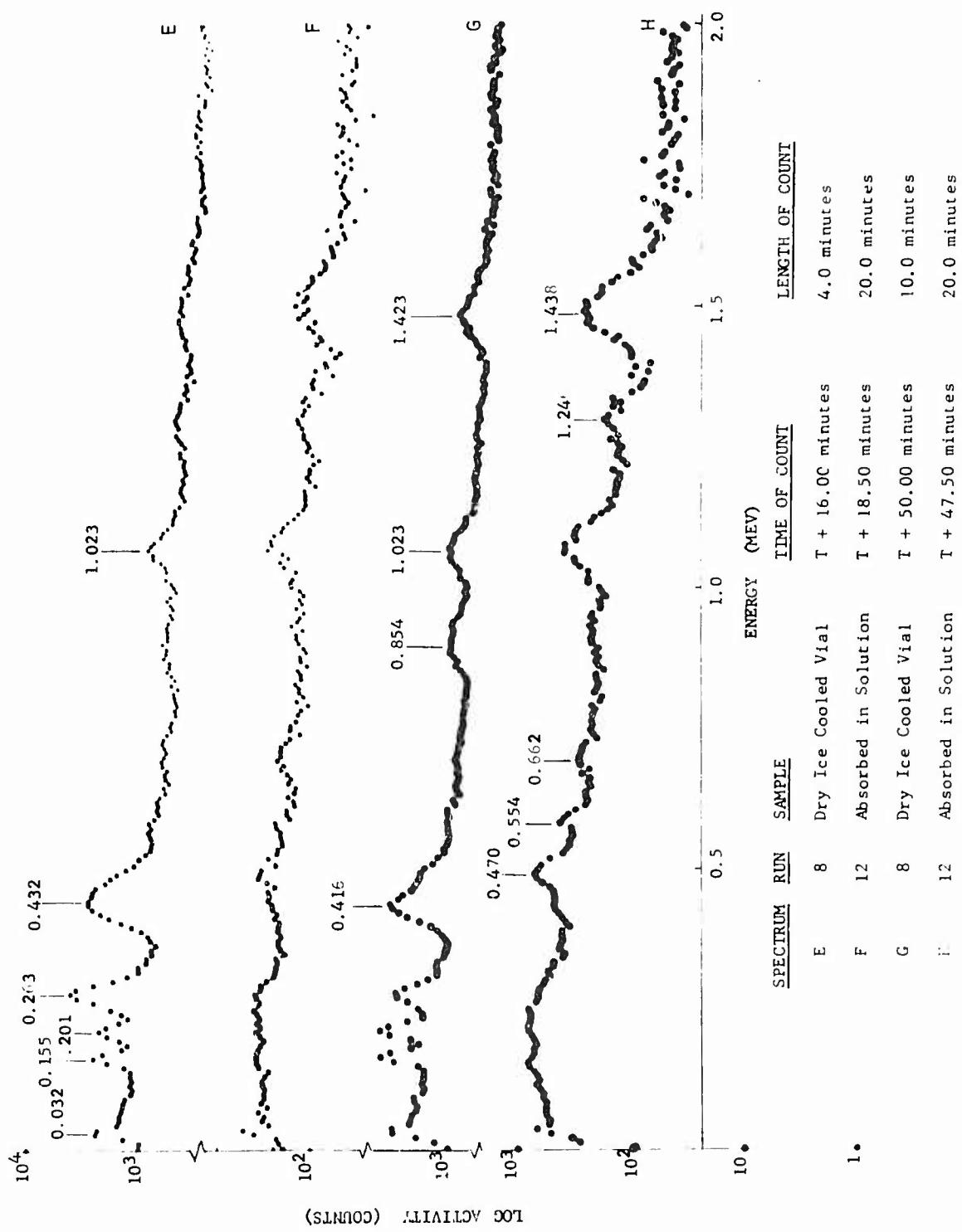
Comparison Spectra of Dry Ice Cooled Gas Sample and Chemically Absorbed Sample.

Figure 4

| SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
E	8	Dry Ice Cooled Vial	T + 16.00 minutes	4.0 minutes
F	12	Absorbed in Solution	T + 18.50 minutes	20.0 minutes
G	8	Dry Ice Cooled Vial	T + 50.00 minutes	10.0 minutes
H	12	Absorbed in Solution	T + 47.50 minutes	20.0 minutes
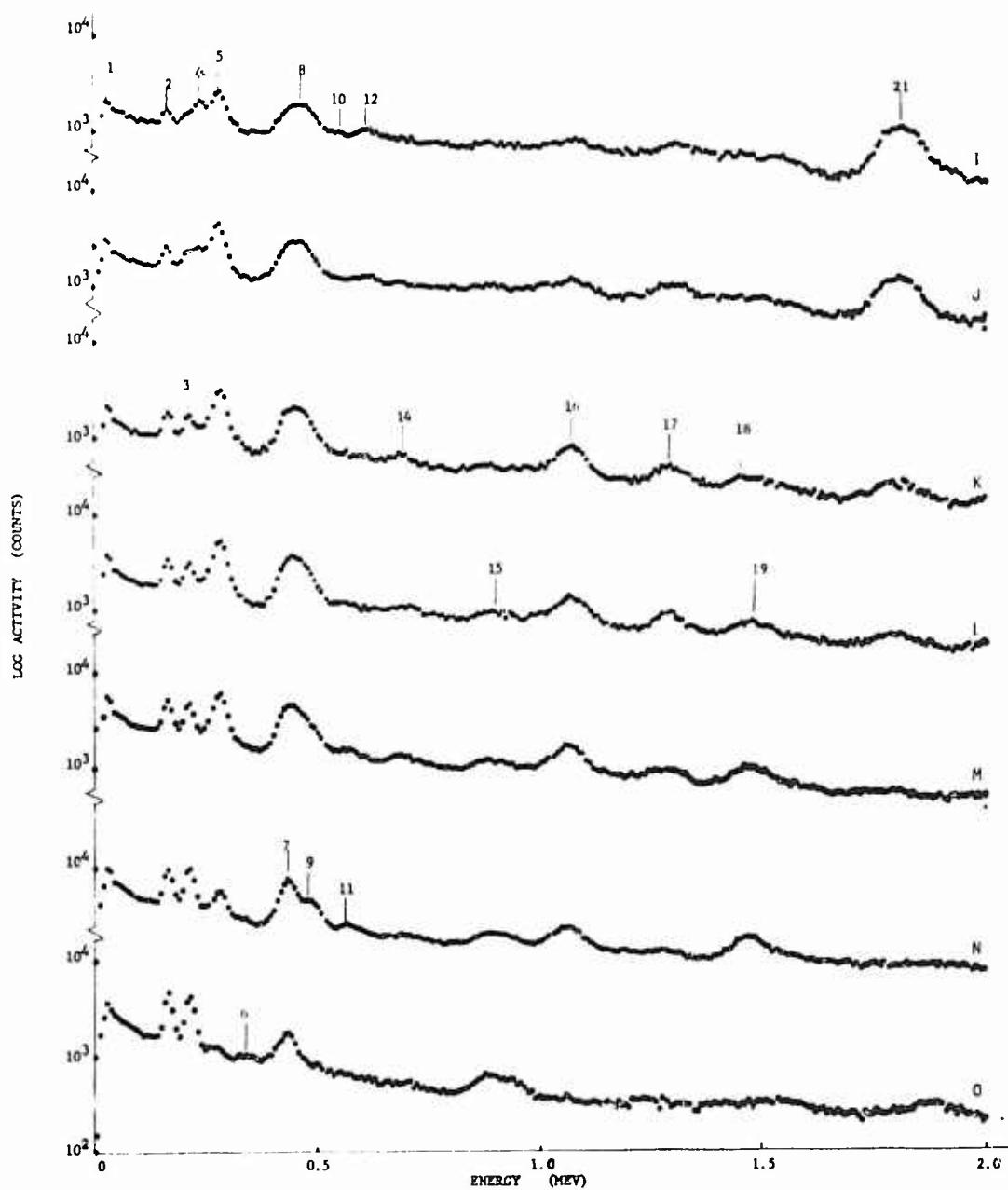
SPECTRUM	RUN	TIME OF COUNT	LENGTH OF COUNT
I	9	T + 2.40 minutes	1.0 minute
J	9	T + 4.75 minutes	2.0 minutes
K	9	T + 11.20 minutes	2.0 minutes
L	9	T + 14.40 minutes	4.0 minutes
M	9	T + 25.00 minutes	8.0 minutes
N	9	T + 50.00 minutes	20.0 minutes
O	9	T + 200.00 minutes	20.0 minutes

Spectrum Decay   0 to 2 MEV Range

Figure 5

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Spectrum Decay 0 to 4 MEV Range

Figure 6

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IV. DISCUSSION

It became apparent through the course of this experiment that
selenium and tellurium isotopes might exist in the radioactive gas
products. Selenium and tellurium could possible be present in the form
of a hydride such as selenium hydride and tellurium hydride which are
gases. Listed in Table III are brief descriptions of each of the
selenium, bromine, krypton, tellurium, iodine, and xenon fission
products that could possibly be identifiable in the reactor gas. A
notation as to photopeak number, and identification by previous
workers is provided. Those fission products of the above elements
which are stable or have a half life that is so long as to make a half
life study impractical are Se^{77}, Se^{78}, Se^{79}, Se^{80}, Se^{82}, Br^{79}, Br^{81},
Kr^{83}, Kr^{84}, Kr^{85}, Kr^{86}, Te^{125m}, Te^{125}, Te^{126}, Te^{127m}, Te^{128}, Te^{129m},
Te^{130}, Te^{131m}, Te^{132}, I^{127}, I^{129}, Xe^{129}, Xe^{131m}, Xe^{131}, Xe^{132},
Xe^{134}, and Xe^{136}. Those fission products whose half life is so short
that indication of their presence is not expected are Se^{77m}, Se^{85}, Se^{87},
Br^{88}, Br^{89}, Br^{90}, Kr^{90}, Kr^{91}, Kr^{92}, Kr^{93}, Kr^{94}, Kr^{95}, Te^{135}, I^{137},
I^{138}, I^{139}, Xe^{140}, Xe^{141}, Xe^{142}, Xe^{143}, and Xe^{144}. Also included
are comments regarding the daughter products such as rubidium and
cesium that may be present. The gamma ray energies and half life of
the isotopes listed below are taken from literature values found in
works by Goldman and Roesser [7], and Der Mateosian and McKeown [8]
who have compiled excellent listings of gamma rays emitted by

radioactive nuclides. The Radiological Health Handbook, published by the U. S. Department of Health, Education and Welfare also contains very useful information on radioactive nuclei [9]. The energies of observed photopeaks may vary somewhat from literature values because of the lack of resolution by the scintillation crystal of two or more gamma rays of similar energies. All of these isotopes decay by beta particle and subsequent gamma emission.

No significance should be made of the third component (See Table II) observed in photopeak 5 (0.263 MEV), because Xe^{135} emits a delayed gamma at 0.25 MEV. This nuclide has been identified by previous workers. The rapid decay of photopeak 21 is caused by the activation of Al^{27} by thermal neutrons to form Al^{28} which then decays by emitting a beta particle and a gamma ray of 1.78 MEV. The half life of Al^{28} is 2.30 minutes, and accounts for the 2.6 minute half life observed in photopeak 21. In a compilation of gamma ray spectra observed in scintillation spectrometry by Heath [10] photopeaks are shown for Al^{28} at 1.78, 1.35, 0.95, 0.40, 0.032 MEV. A gas sample was obtained in which all normal procedures were followed with the exception that the fissionable fuel elements were not placed in the aluminum cylinder. The resulting spectrum showed a strong photopeak at 1.75 MEV which decayed away within twenty minures after reactor shut down. The chemical form of the aluminum in the gas is probably an aerosol suspension of aluminum oxide particles.

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POSSIBLE NUCLIDES DETECTABLE IN EARLY GAS SPECTRA

Isotope	Half Life	Detectable Photopeaks Principle	Secondary	Observable in Early Spectra *	Observed Half Life **	Previous Identification
Se^{79m}	3.9 min	0.096		None		
Se^{81m}	56.8 min	0.103		None		
Se^{81}	18 min	0.28		5	15.6 min	
Se^{83m}	69 sec	1.01	2.02	None		
Se^{83}	25 min	0.173	0.368	3 & 4	22.1 min	
Se^{81}	3 min	No Gamma Energies Reported				
Br^{83}	2.4 hrs	0.051		None		
Br^{84m}	6 min	0.88	1.46	None		
Br^{84}	31.8 min	0.879	1.90	None		
Br^{85}	3.0 min	0.81	0.92	None		
Br^{86}	54 sec	1.6	1.4	None		
Br^{87}	55 sec	1.4	2.6	None		
Kr^{85m}	4.36 hr	0.1495	0.304	2 & 6	3.79 hr	[1], [3], [4]
Kr^{87}	76 min	0.403	1.75	7	75.1 min	[1], [3], [4]
				21	81.5 min	
Kr^{88}	2.77 hr	0.191		3 & 4	2.14 hr	[1], [3], [4]
Kr^{89}	3.2 min	0.60	0.22	12	2.56 min	[4] ***
				4	3.39 min	#
Rb^{88}	17.8 min	1.853	0.906	22 & 15	Daughter of Kr^{88}	[3] ***, [4] (See Fig. 5)
Rb^{89}	15.4 min	1.05		16	Daughter of Kr^{89}	[3] ***, [1] *** (See Fig. 5)
Te^{127}	9.3 hr	0.418	0.360	7		
Te^{129}	57.3 min	0.0268	0.475	1 & 9		
Te^{131}	24.8 min	0.15	0.448	2	18.2 min	
				9	27.7 min	
Te^{133m}	50 min	0.334		None		
Te^{133}	2 min	0.6	1.0	None		
Te^{134}	42 min	0.030		1	##	
I^{131}	8.05 day	0.364		None		[3], [4]
I^{132}	2.26 hr	0.677	0.778	None		
I^{133}	20.8 hr	0.54		None		[3] ***, [4] ***

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Isotope	Half Life	Detectable Principle	Photopeaks Secondary	Observable in Early Spectra *	Observed Half Life **	Previous Identification
I^{134}	5.2.5 min	0.86		None		[4] ***
I^{135}	6.7 hr	1.14	1.275	None		[3] *** [4] ***
I^{136}	83 sec	1.32		None		[4] ***
Xe^{133m}	2.35 day	0.233				[1] [4] [4]
Xe^{133}	5.27 day	0.0809				[1] [4] [4]
Xe^{135m}	15.3 min	0.53		12	16.1 min	[4] ***
Xe^{135}	9.2 hr	0.249		5		[1], [4] [?]
Xe^{137}	4.2 min	0.44		7 & 8	2.76 min	[3] ***
				8 & 9	2.56 min	
Xe^{138}	17 min	0.42		7	14.3 min	[4] [4]
Xe^{139}	41 sec	0.22		3	33.6 sec	
Cs^{137}	30 yr	0.662			Daughter of Xe^{137}	###
Cs^{138}	32.2 min	1.436		19	Daughter of Xe^{138}	[4], [4]
Cs^{139}	9.5 min	1.28	0.63	17 & 14	Daughter of Xe^{139}	

*   Photopeak numbers from Table I and Figures 4 and 5.

**   Half Life values from Table II and averaged where appropriate.

***   Photopeak identification with no supporting half life data.

\#   5.13 min half life in photopeak 3 may be caused by interference from this isotope.

\##   This isotope is probably the major contributor to photopeak 1. Other contributions from Te^{129} and from low energy gamma rays and x-rays from other isotopes are likely.

\###   This isotope is not observable in early spectra but is detectable in the background spectra at the reactor facility. This species has probably built up through out the laboratory by long term leakage from the reactor control rods.

TABLE III

V. CONCLUSIONS

Radioactive nuclides present in the gaseous discharge from the uranium fuel of the AGN-201 reactor at the Naval Postgraduate School are summarized in Table IV. The technique employed is adequate to obtain a gas sample for the determination of half lives between 0.5 and 20 minutes. The absence of the strongest photopeaks for bromine and iodine gases in the initial spectra suggests the bromine and iodine fission products are largely trapped in the matrix of the uranium-polyethylene fuel. While there may be trace quantities of these halogens present in the gas, they cannot be identified.

NUCLIDES IDENTIFIED

Initial Volatile Nuclides:

Te^{131}

Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{135}, Xe^{137}, Xe^{138}, Xe^{139}

Se^{81}, Se^{83}

Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, Kr^{89}

Active Daughters:

Cs^{137}, Cs^{138}, Cs^{139}

Rb^{88}, Rb^{89}

I^{131}

Nuclides Possibly Present*

I^{132}, I^{133}, I^{134}, I^{135}, I^{136}

Te^{127}, Te^{129}, Te^{134}

Br^{83}, Br^{84m}, Br^{84}, Br^{85}, Br^{86}, Br^{87}

*Based on photopeak analysis with no supporting half life data.

TABLE IV

APPENDIX A

EQUIPMENT

1. Counting Chamber: A 2-inch thick lead cube, 22 inches square
    by 28 inches high, lined with 1/8-inch
    aluminum and 1/4-inch high copper content
    brass.

2. Scintillation Detector: Harshaw 12SW12-W3, 3-inch by 3-inch
    cylindrical well type NaI (Th) crystal with
    matched photomultiplier tube.

3. 512 channel multichannel analyzer, consisting of the following
    components:

    a. Nuclear Data, Inc. ND-180F Analog to Digital Converter

    b. Nuclear Data, Inc. ND-180M 512 Channel Memory Unit

    c. Nuclear Data, Inc. ND-180R Read out Control Unit

    d. Textronix, Inc. Rm503 Oscilloscope

    e. Tally Corp. Model 1506 Reader/Perforator

    f. Teletype Corp. Teletypewriter

    g. Hewlett-Packard, Co., Mosley 7590 CR Automatic
        Plotting System

4. Single channel analyzer (2 ea): Hewlett-Packard Model 5201L
    Scaler-Timer

5. Mercury Switch Pulser: Hamner Model NP - 10.

6. Vacuum System: A glass vacuum system with a CENCO HYVAC
    2 pump and an oil diffusion pump. Pressure measurement:
    Thermocouple gauge calibrated to 1 micron. (Pressures actually
    used were lower than this and were estimated to be 10^{-4} mm when
    the meter was "pegged").

7. Reaction Vessel: A cylindrical aluminum contained approximately
    2 x 5 inches fitted with a vacuum stainless steel valve.

8. Fuel: 8 grams of UO_2, 20% enrichment in U^{235}, in approximately
    200 grams of polyethylene.

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13 ABSTRACT

The volatile radioactive nuclides produced by thermal neutron fissioning of uranium oxide-polyethylene fuel of the AGN-201 Reactor were collected in an evacuated aluminum cylinder. The following constituent isotopes, identified by gamma ray spectrometry, present in 20 minute old gas are; Te^{131}, Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{137}, Xe^{138}, Xe^{139}, Se^{81}, Se^{83}, Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, and Kr^{89}. Radioactive daughter products identified are Cs^{137}, Cs^{138}, Cs^{139}, Rb^{88}, Rb^{89}, and I^{131}. Other isotopes of iodine, tellurium, and bromine may be present. Spectra were taken with a NaI (Th) scintillation crystal.

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KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
gamma-ray spectrum						
fission products						
gas						
selenium						
bromine						
krypton						
tellurium						
iodine						
xenon						
isotopes						